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Reduction in surface recombination and enhancement of light emission in silicon photonic crystals treated by high-pressure water-vapor annealing

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We propose and demonstrate the application of high-pressure water-vapor annealing (HWA) to silicon photonic crystals for surface passivation. We find that the photoluminescence intensity from a sample treated with HWA is enhanced by a factor of ~ 6 . We confirm that this enhancement originates from a reduction in the surface-recombination velocity (SRV) by a factor of ~ 0.4 . The estimated SRV is as low as 2.1×10^3 cm/s at room temperature. These results indicate that HWA is a promising approach for efficient surface passivation in silicon photonic nanostructures. © 2010 American Institute of Physics. [doi:10.1063/1.3489419]

Photonic nanostructures, including photonic crystals (PCs), in which the refractive index varies periodically on a length-scale comparable to the optical wavelength of interest, have attracted much attention as a powerful tool for the manipulation of photons.^{1,2} However, the influence of the surface is significant in photonic nanostructures due to the large surface-area-to-volume ratio. Consequently, controlling surface recombination (SR) is of importance in active photonic-nanostructure devices,³ including light emitters, receivers, and switches, which use excited carriers. The SR on photonic nanostructures can be reduced through the introduction of quantum nanostructures,⁴ plasma irradiation,⁵ and chemical treatment⁶ in the direct band gap compound semiconductors that are currently the dominant material in active devices.

Recently, there have been several reports on the introduction of PCs (Refs. 7 and 8) and nanocavities^{8–11} into silicon (Si) as emitters, as a step toward future integration with electronic devices. Although light emission has been observed, the efficiency has been restricted by significant SR. If the SR could be suppressed, efficient emission might be achieved because the development of Si-based PC devices is progressing rapidly.^{12–21} However, to date, there have been no reports of the successful passivation of the surface of Si-based photonic nanostructures.²²

Here we propose the application of high-pressure water-vapor annealing (HWA) (Ref. 23) to Si PCs. We have previously reported that the light emission of Si quantum nanostructures such as nanocrystalline porous Si (nc-pSi) (Ref. 23) and Si nanowires,²⁴ the dimensions of which are much smaller than those of photonic nanostructures, is greatly enhanced by HWA.

Here we report on the emission spectra and time-resolved photoluminescence (PL) measurements of Si PCs treated by HWA. The reduction in the surface-recombination

velocity (SRV) caused by HWA is evaluated.

We employed a two-dimensional PC slab with a triangular lattice of air holes.⁸ The lattice constant (a) was varied from 290 to 1000 nm. We also fabricated a tuned L3 PC cavity,¹² comprised of a line of three filled holes with the holes at either end being shifted toward the edges by $0.1a$. The samples were fabricated from a Si-on-insulator (SOI) wafer by electron-beam lithography and inductively coupled plasma etching. The thickness and resistivity of the p-type boron-doped Si slab situated above the $3 \mu\text{m}$ SiO_2 layer were 220 nm and $\sim 20 \Omega \text{ cm}$, respectively. The freestanding slab was formed by selective chemical etching of the SiO_2 layer on the Si substrate using hydrofluoric acid solution. Subsequently, for the HWA, the samples were placed into a container with a specified quantity of deionized water, and heated at 260°C for 3 h. From the results for nc-pSi,²⁵ a water-vapor pressure of between 1.3 and 3.9 MPa is suitable to obtain significant defect reduction and stress release. These effects saturate with time, and 3 h were shown to be enough for nc-pSi.²⁵ At a temperature of 260°C , a maximum pressure of ~ 4 MPa can be achieved, corresponding to the water saturation pressure. However, when the pressure was set to 3.9 MPa, atomic-force microscopy revealed that HWA induced a surface roughness of 4 nm due to too strong oxidation. In contrast, no HWA-induced surface roughness was observed for a pressure of 1.3 MPa. The estimated surface roughness of 0.2 nm was similar to that of an unprocessed SOI wafer. Finished samples had uniform air holes, as shown in the inset of Fig. 1.

The PL spectra were measured by a gallium indium arsenide detector array with a monochromator through an objective lens with a numerical aperture of 0.95 at room temperature (RT). The wavelength and spot size of the continuous-wave (cw) pumping laser were 488 nm and $\sim 2 \mu\text{m}$, respectively. Six clear resonant peaks from the cavity in the PL spectrum within the Si band-edge emission wavelength were observed (Fig. 1).²⁶ The wavelengths of these peaks were in good agreement with a finite-difference

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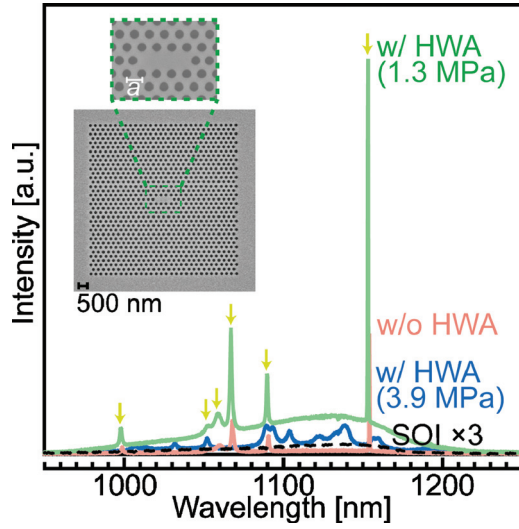


FIG. 1. (Color) PL spectra. Black dashed line denotes original SOI and red denotes cavity without HWA. Green and blue denote HWA-treated cavities with process pressures of 1.3 MPa and 3.9 MPa, respectively. Arrows indicate cavity modes. Inset shows a scanning electron micrograph of a fabricated sample with HWA (1.3 MPa).

time-domain simulation of the L3 PC cavity.⁸ The sample treated with HWA at the process pressure of 1.3 MPa enhanced the efficiency by a factor of ~ 6 . The resonant wavelength of the sample was blueshifted by ~ 1 nm, due to a reduction in the effective refractive index by the HWA-induced surface oxidation. In contrast, the resonant peaks from the sample treated with HWA at 3.9 MPa were deformed due to the generated surface roughness.

Figure 2 shows the spectrally integrated PL intensity (L), which represents the state of the carriers in the Si, against the irradiated power (I) of the Si PC with no cavity at RT. The intensity L was roughly proportional to I^2 for both HWA-treated (1.3 MPa) and untreated samples. The difference in L between samples was a factor of ~ 6 .

The rate equations for the carrier densities N can be written as²⁷

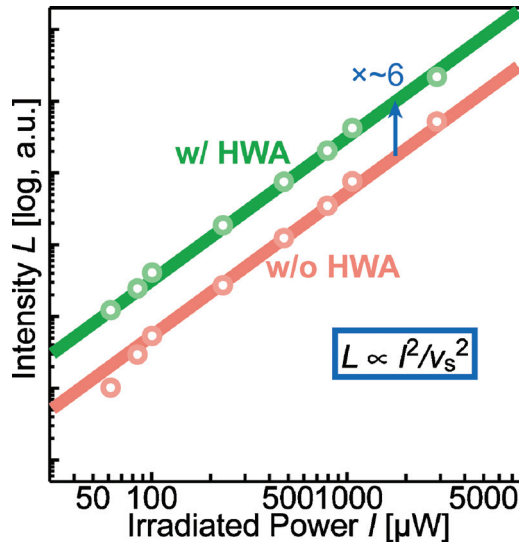


FIG. 2. (Color) L vs I of Si PC without cavity. Experimental plots show fit where L is proportional to I^2/v_s^2 . Red and green data denote samples without and with HWA, respectively.

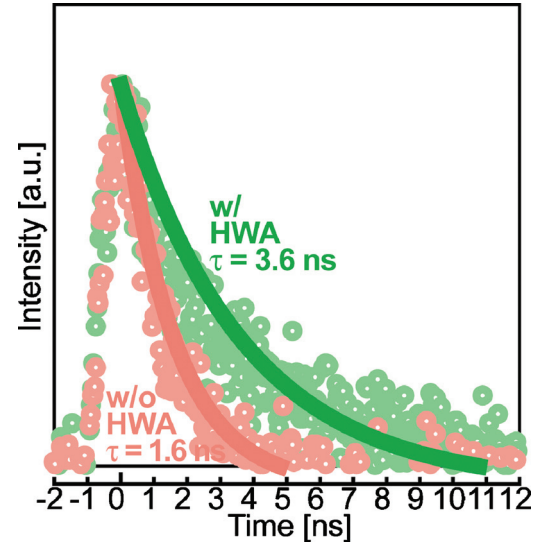


FIG. 3. (Color) Time-resolved PL of Si PC. Green and red plots denote samples with and without HWA, respectively. Solid lines show fit using a single exponential function to estimate τ .

$$\frac{dN}{dt} = \frac{\eta_a}{V_{ex}E_{ex}}I - \left[\frac{1}{\tau_{nr}} + BN \right]N, \quad (1)$$

where η_a is the absorption ratio of the pumping light in the Si, V_{ex} is the pumped volume, E_{ex} is the photon energy of pumping light, τ_{nr} is the carrier lifetime of the nonradiative process, and B is the radiative-recombination coefficient. The nonradiative lifetime can be expressed as

$$\tau_{nr}^{-1} = \tau_{nr0}^{-1} + \tau_{sf}^{-1} = \tau_{nr0}^{-1} + v_s \times A/V, \quad (2)$$

where τ_{nr0} is the intrinsic nonradiative-recombination lifetime including Shockley–Read–Hall recombination and the Auger process, τ_{sf} is the SR lifetime, v_s is the SRV, A is the surface area, and V is the volume of the sample.²⁸ The intensity of the observed emission can be expressed as $L = \eta_e BN^2$, taking into account the light-extraction efficiency of the measurement system (η_e). Thus, the emission L should be proportional to I^2/v_s^2 , if the SR process is dominant ($\tau_{nr}^{-1} + BN \sim v_s \times A/V$). This relationship indicates that the v_s in the Si PC is reduced by a factor of ~ 0.4 ($=1/\sqrt{6}$) upon HWA.

In order to estimate v_s , we performed time-resolved PL measurements at RT using a streak camera. We employed a laser diode with a pulse width of ~ 40 ps, a repetition frequency of 1 MHz, an excitation wavelength of 403 nm, a spot diameter of ~ 6 μm , and an irradiated energy of 5 pJ/pulse. The results are shown in Fig. 3. The decay curve of a sample ($a=800$ nm) treated with HWA was slower than that without HWA. The carrier lifetimes of the samples with and without HWA were 3.6 ns and 1.6 ns, respectively. These are much shorter than the typical lifetimes ($> \sim \mu\text{s}$) of the radiative emission and Auger processes in crystalline Si.²⁹ Therefore, we conclude that the present carrier lifetime was largely determined by the SR.

The inverse carrier lifetime due to SR, τ_{sf}^{-1} , can be expressed as shown in Eq. (2). By varying a in the PC, the surface-area-to-volume ratio (A/V) can be changed. As a result, v_s can be estimated experimentally²⁸ from the gradient of the fitted line, as shown in Fig. 4. We obtained $v_s=4.8 \times 10^3$ cm/s for the sample without HWA. This is in good

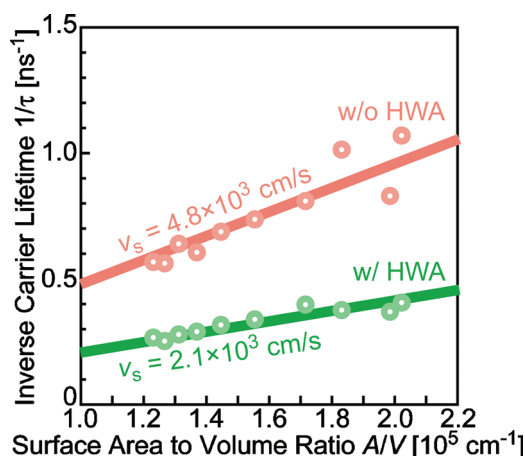


FIG. 4. (Color) Estimation of experimental v_s , calculated by plotting $1/\tau$ vs A/V . Green and red denote samples with and without HWA, respectively.

agreement with the value reported for bulk Si without any passivation processes.³⁰ In contrast, v_s for the sample with HWA was as low as 2.1×10^3 cm/s. The change in v_s by a factor of ~ 0.4 caused by HWA is consistent with the value predicted from Fig. 2, as discussed above. These results show that the HWA efficiently suppresses the nonradiative centers by surface passivation, forming a high-quality surface oxide at a low treatment temperature (260 °C), as was also the case for nc-pSi.²⁵ The reduction in SRV by a factor of ~ 0.4 is comparable to the change in the inverse carrier lifetime of nc-pSi on HWA (1/2–1/3).²⁵

If a high-quality oxide is formed, additional strain should not occur in the Si upon HWA. In order to investigate strain in the Si PC, we performed Raman microspectroscopy³¹ using a 488 nm cw laser. Figure 5 shows the Raman spectra for the samples with and without HWA (1.3 MPa). No significant change in the spectra occurred, demonstrating that no strain was generated by the HWA. A further reduction in the SR would be expected upon further optimization of the treatment conditions, as an external quantum efficiency of $>20\%$ has been achieved in nc-pSi under appropriate conditions.²³

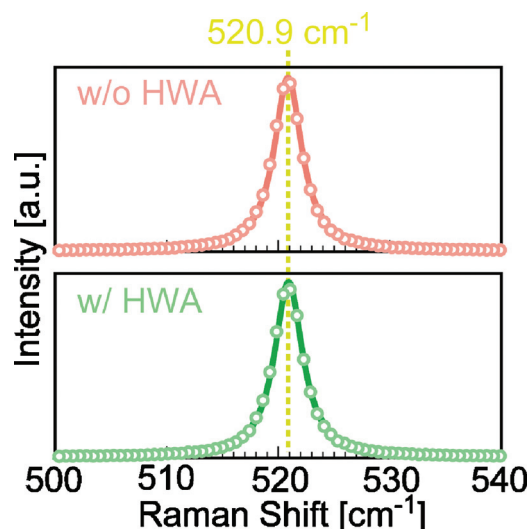


FIG. 5. (Color) Raman spectra at RT. Green and red plots denote samples with and without HWA, respectively. Solid lines show fit using a Lorentzian function.

In summary, we have applied HWA for the surface passivation of Si PCs. We found that HWA enhances the light-emission intensity and reduces the SRV. Our results demonstrate that HWA is a promising approach for producing highly-efficient Si photonic-nanostructure devices.

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